

Irreversibility in Ising and Heisenberg spin glasses (invited)

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Spin glasses are rather unique systems which show simultaneously apparent phase transitions as well as metastable or glassy behavior. The existence of irreversible behavior appears to be intimately connected with the phase transition. Here, we will review experiments which show irreversible phenomena and will deduce from the data a simple heuristic picture of the free energy surface $F[m_i]$, where m_i is the thermally averaged spin at site i . The picture that follows from this analysis is then made more rigorous within a calculational scheme, in which, for large-size systems, we numerically compute the evolution of minima of mean field models for $F[m_i]$ as the magnetic field H and temperature T are changed. For Ising spins, magnetic hysteresis, field-cooled, zero-field-cooled, and remanent magnetizations are computed and found to be in good qualitative agreement with experiment. For Heisenberg spins, we find no irreversibility unless anisotropy is present. We discuss the re-entrant ferromagnet-spin glass transition as well as the effects of various kinds of anisotropy, on vector spin glasses. The overall good qualitative agreement between theory and experiment lends support to our hypothesis that, on intermediate time scales, the behavior of spin glasses reflects the properties of the free energy surface: that irreversibility occurs when minima of F are destroyed with changing H or T .

PACS numbers: 75.30. — m, 05.70. — a, 64.60.Cm

I. INTRODUCTION

In the last few years theoretical and experimental studies of spin glasses have been increasingly concerned with nonequilibrium and time-dependent effects. Early theoretical treatments¹ were based on the presupposition that a thermodynamic phase transition existed. However, shortly thereafter, experiments²⁻⁶ provided clear evidence that at low temperatures the dc magnetization and other properties were time and history dependent. Therefore an equilibrium, thermodynamic approach seems to be inappropriate. This failure of equilibrium thermodynamics is presumably connected to the breakdown of ergodicity. The spin glass does not sample all accessible states in the time scale of a laboratory experiment.

There have been a number of theoretical approaches aimed at dealing with these nonequilibrium effects. These have built on either dynamical^{7,8} or replica symmetry breaking⁹⁻¹¹ concepts. Our own work^{12,13} has focused on nonergodicity in spin glasses and is based on the assumption that for intermediate time scale experiments the spin glass is contained within a local minimum of the free energy surface. As the field or temperature changes, this surface evolves; the spin glass follows the evolution of a given minimum as long as it persists and when it disappears, the system hops to a nearby minimum in which it is trapped for some period of time. The results obtained numerically by following the evolution of the free energy surface yield various history-dependent magnetizations which are in good qualitative agreement with experiment. In this paper we will review the essential aspects of this work.

As will be seen below, our techniques are amenable to a treatment of Heisenberg as well as Ising spin glasses. In the

Heisenberg case, we have been able to study the effects of various kinds of anisotropy on the history-dependent magnetizations for large-size systems.¹³ Within conventional simulation techniques such studies have not been feasible, because of computational limitations. Of particular interest in the last few years has been the phenomenon of "re-entrant" spin glass behavior, which occurs in (presumably) anisotropic Heisenberg spin glasses which are close to the critical concentration for ferromagnetism. For these systems it appears that there is long-range ferromagnetic order at high temperatures which is destroyed at lower T when a spin-glass-like state sets in.¹⁴ While experiments¹⁵ cannot unequivocally prove that the ferromagnetic order is of truly infinite range, it is nevertheless clear from a variety of different measurements^{16,17} that in these alloys there is some kind of "transition" to a new low-temperature state. Furthermore at low temperatures there appears to be a considerable amount of irreversibility which is a signature of spin glass behavior.¹⁷

As yet, no theoretical studies have provided a convincing explanation of this re-entrant behavior; we summarize here our numerical work which demonstrates that Heisenberg spin glasses with conventional types of temperature-independent anisotropy do not appear to undergo a re-entrant transition.

II. EXPERIMENTAL DATA

One of the first indications of the unusual properties of spin glasses was the observation of their history-dependent magnetization.²⁻⁶ The zero-field-cooled (zfc) magnetization is obtained by cooling the sample to the measuring temperature in zero external field starting at high temperatures; then a field H is applied and M is measured. A second field-cooled

(fc) magnetization is obtained by turning on the field H above T_c and then cooling at constant $H \neq 0$. In general, the zfc and fc M differ for $T < T_c$. For some systems, e.g., CuMn, the zfc M is independent of the measuring time³ for $T < T_c$, while for other systems, e.g., AuFe, the zfc magnetization changes slowly with time⁶ for T below T_c ; for long times its value approaches the fc value. Because of the time independence of the fc magnetization, it is believed that the most likely candidate for the true equilibrium state is that obtained upon cooling at constant field. On heating, the fc magnetization is reversible, whereas the zfc magnetization is not. It is important to note that if a zfc magnetization is cooled back down (sufficiently below T_c), a reversible and relatively T -independent magnetization is obtained.

There exists an irreversibility temperature above which $M^{fc} = M^{zfc}$. A plot of this characteristic irreversibility temperature versus field was found experimentally⁴ to be similar to the phase diagram suggested by deAlmeida and Thouless,¹⁸ based on theoretical arguments. For T higher than the deAlmeida–Thouless line the system is reversible and history independent, while below the line, M depends on the sample history. Note that the larger the applied H , the lower T one can go before irreversibility sets in.

An alternative way of representing history-dependent effects is to study M vs H at fixed T . Experimental studies^{19–21} show that the shape of the hysteresis curve varies from one spin glass alloy to another. An additional manifestation of irreversibility is the existence of remanent magnetization. The various remanences show an initially rapid decrease with time t followed by a much slower (often $\propto \ln t$) decay. From these data, we can draw the following conclusions:

- (1) Cooling processes are reversible. Both the fc magnetization and the partially cooled section of the zfc magnetization are evidence for this observation.
- (2) Heating processes are generally irreversible. This can be seen by cooling the zfc magnetization (obtained upon warming).
- (3) Changing H is irreversible for all but very large H . This irreversibility is manifested by the existence of hysteresis loops. Irreversibility occurs even for extremely small⁴ ΔH .
- (4) There are two rather distinct time scales in the data.²² (i) rapid relaxation ($t \sim 10^{-11}$ sec) and (ii) slow relaxation $\sim \ln t$ ($t \gtrsim 10^{-7}$ sec).

III. FREE ENERGY SURFACE

These four general properties of the data provide a heuristic picture of the free energy surface $F[m_i]$. We define $m_i = \langle S_i \rangle$, where $\langle \rangle$ stands for a thermal average. The irreversibility and metastability of spin glasses naturally arises if there are many states in which $F[m_i]$ is locally stable. All that is needed to explain the data are the following features of $F[m_i]$, which we will show below to be a consequence of a more detailed calculational scheme.^{12,13}

- (1) A minimum in the free energy at temperature T persists to all lower T .
- (2) Below T_c , the number of minima increases as T decreases.

(3) Minima disappear with small changes in H (and reappear in other regions of $F[m_i]$).

In general, irreversibility is connected with the disappearance of minima with H or T .

To understand how irreversibility is related to the free energy surface, we presume that there exists an “intermediate” time scale, such that the system can relax to and be contained in the nearest free energy minimum. The time necessary to find the true ground state is very long, since the system must presumably minima hop between many metastable states, which are separated by large barriers. This accounts for the slow relaxation processes. The fast processes, on the other hand, are thought to correspond to the small rearrangements of spins which must occur when the minima are deformed as H or T changes. In order for the nature of the free energy surface to be relevant for considering irreversible processes, we must assume that on an intermediate time scale the spin glass “follows” a minimum of the free energy surface as it evolves in H or T .

Let us now put these ideas on a firmer footing. As in most theoretical descriptions of spin glasses, we consider a Hamiltonian in which spins are located at all sites and the nearest-neighbor exchange interaction J_{ij} is given by a Gaussian probability distribution $P(J_{ij})$ of width \bar{J} and mean J_0 . In what follows, T, H, J_0 , etc. will be measured in units of \bar{J} ; we also take the magnetic moment $g\mu_B = 1$. The general free energy functional $F[m_i]$ can be decomposed into the mean field [mf] and so-called reaction [reac] terms, where for general S , such that $-S < m_i < S$,

$$F^{\text{mf}}[m_i] = \frac{1}{2} \sum_{ij} J_{ij} m_i m_j - k_B T \sum_i \ln \frac{\sinh(S + 1/2)H_i}{\sinh H_i/2}. \quad (1)$$

Here $H_i = \beta \sum_j J_{ij} m_j + \beta H$, and $\beta = 1/k_B T$. For the infinite Ising spin 1/2 model, Thouless, Anderson, and Palmer¹ (TAP) have shown that

$$F^{\text{reac}}[m_i] = -\frac{1}{2} \beta \sum_{ij} J_{ij}^2 (1 - 4m_i^2)(1 - 4m_j^2). \quad (2)$$

In general, this term derives from the fact that a “self-orientation” effect, present in F^{mf} , should be cancelled out.

A number of groups^{23–25} have searched for minima of the TAP free energy surface for small systems, $N \lesssim 100$. Generally, solutions are found only in about 10% of all bond configurations and those that are found quickly disappear as the temperature is varied. It is now believed that the reaction term given in Eq. (2) must be corrected in small-size systems. However, at the present time there is no fully consistent theory of these corrections.

Because a characterization of the spin glass free energy appears to be so important to our understanding of these systems, we have studied the more well-behaved mean field limit. It should be recalled that at very low and high temperatures the TAP self-consistent equations are equivalent to those derived from F^{mf} . In contrast to the TAP case, there are no unphysical extrema: the entropy is always well behaved.

We solve iteratively the self-consistent equations deriving from $\partial F^{\text{mf}} / \partial m_i = 0$. We consider $[m_i]$ to be converged,

when

$$\frac{\sum_i [m_i^n - m_i^{n-1}]^2}{\sum_i (m_i^n)^2} < 10^{-6}, \quad (3)$$

where the superscript n denotes the n th iteration. After a solution is found at a particular H and T , we follow the minimum with H or T , using the previously converged values as the initial guess. It is important to note that iteration generates only minima. We have verified that minima are not lost due to numerical artifacts. We always varied H and T according to the experimental prescription and in this way generated fc and zfc magnetizations as well as hysteresis loops.

In addition to Ising spin systems, it is rather easy to study vector spin models as well using this method. This case is of considerable interest, since it allows us to study the effect of anisotropy. The anisotropy terms in the Hamiltonian are given by

$$H^A = -\sum_i D (S_i^z)^2 - \sum_{ij} D'_{ij} (S_i \times S_j). \quad (4)$$

Here the first and second terms correspond to uniaxial and Dzyaloshinsky-Moriya²⁶ (DM) anisotropy, respectively. The limit $D \rightarrow -\infty$ corresponds to the x - y model and $D \rightarrow \infty$, the Ising model. We assume that the coefficients $D'_{ji} = -D'_{ij}$; $P[D'_{ij}] = \delta(D'_{ij} \pm D')$. The parameters D and D' are chosen as variable parameters, in our numerical studies. The generalizations of the mean field equations for the Heisenberg model are straightforward and given in Ref. 13.

Within the context of our mean field theoretic calculation, fluctuation effects are ignored. The spin glass is assumed to "sit" at the bottom of a given well as long as it remains a minimum and, otherwise, to hop into a nearby state. We expect that the rapid relaxation observed in spin glasses may be associated with a rapid relaxation to the nearest minimum; the slow relaxation processes are related to the very much slower thermal activation processes which carry the system from one minimum to another. Our mean field approach is designed to treat only those experimental measurements which have very slow time dependences ($\sim \ln t$), so that the system is always "quasi-equilibrated."

Based on our numerical work, our most general observations, about the way in which the free energy evolves with H or T , for Heisenberg and Ising spins are as follows:

- (1) A free energy minimum never disappears upon cooling.
- (2) Below $T_c(H)$, a minimum will generally disappear upon heating (unless the minimum was obtained by a cooling procedure),
- (3) For $T < T_c(H)$, minima appear to be continuously created as well as destroyed, upon changing H , by small but finite amounts.

IV. ISING MODEL RESULTS

In both the Ising and Heisenberg cases zero-field-cooled states were obtained by cooling in zero H from high $T > 1.5T_c$ to $T \sim 0$. At $H = 0$ we define $T_c \equiv T_c(0)$ as the lowest temperature at which $Q \equiv N^{-1} \sum_i m_i^2$ extrapolated to the thermodynamic limit is nonzero. To measure M , a field was applied after cooling to the lowest T . The T -dependent magnetization M^{zfc} is then obtained by warming from low T

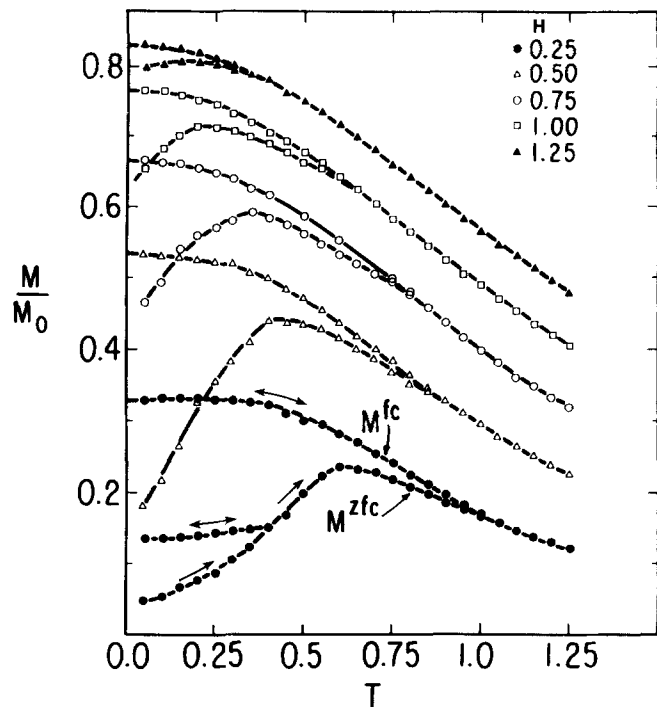


FIG. 1. Field-cooled (upper curves) and zero-field-cooled (lower curves) magnetizations vs temperature T/J for various H/J for the $2d$ Ising model for $J_0 = 0$, $N = 30^2$, and $S = 1/2$. M is normalized by $S = 1/2$.

as in the experimental procedure. In Fig. 1, the $2d$ results are shown for $S = 1/2$ Ising model with $J_0 = 0$. It is important to realize that after the field is applied and the temperature raised, a subsequent cooling leads to irreversibility. Also shown is the field-cooled magnetization obtained upon cooling at constant field $H = 0$, M^{fc} . These fc states were always found to have the lowest free energy for a given (H, T) . They were also completely reversible with temperature.¹²

Our results are in qualitative agreement with experimental data although finite-size effects tend to blur out the transition region so that M^{fc} and M^{zfc} do not meet precisely at the maximum in M^{zfc} .

V. HEISENBERG MODEL RESULTS

A. Isotropic model

We have extended the analysis discussed in the previous section to the Heisenberg model, with and without anisotropy. As first reported elsewhere,¹³ we have found that the short-range, isotropic Heisenberg Hamiltonian has no macroscopic irreversibility. This is presumably because of the accessibility of the fc state which has an infinite number of degeneracies each corresponding to the rotations about the field axis.

In the Heisenberg case in zero H , the Edwards Anderson order parameter is directionally dependent when a spontaneous magnetization is present:

$$Q_{\perp} = \frac{1}{2N} \sum_i (m_i^x)^2 + (m_i^y)^2,$$

and

$$Q_{\parallel} = \frac{1}{N} \sum_i (m_i^z)^2,$$

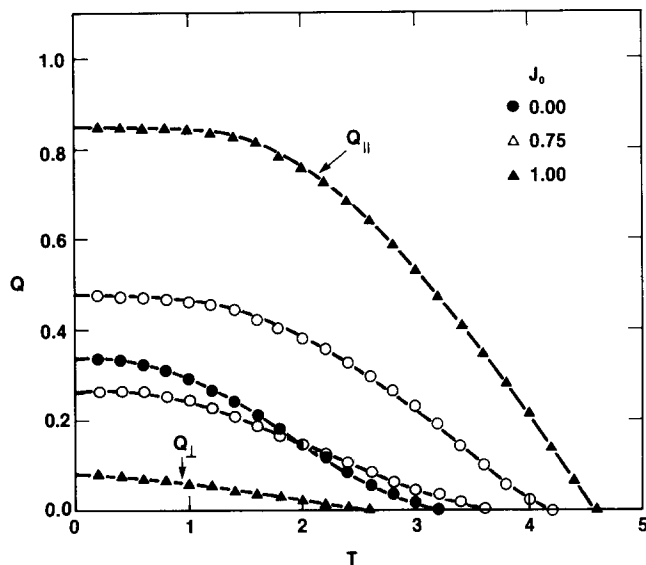


FIG. 2. Temperature dependence of the spin glass order parameters ($Q_{||}$ and Q_{\perp}) for the isotropic Heisenberg model with different values of J_0 , for $N = 10^3$ and $S = 1$.

where z is the direction of the spontaneous magnetization. In Fig. 2 we plot these order parameters as a function of T for various J_0 . At the multicritical point at which the ferromagnetic, spin glass, and paramagnetic phases meet, $J_0 \equiv J_{oc} \sim 0.55$. Above this J_0 , $Q_{||}$ becomes nonzero at a higher temperature than does Q_{\perp} . The onset of $Q_{||}$ corresponds to the onset of ferromagnetism, whereas when Q_{\perp} becomes nonvanishing, ferromagnetic and spin glass order will coexist. For J_0 about ~ 1.25 , Q_{\perp} is completely suppressed.

Since our mean field calculation is exact in the vicinity of $T = 0$, we can reliably calculate the shape of the phase diagram for Heisenberg spin glasses at low T . (Additionally we have checked our numerical procedure by comparing the Ising phase diagram with analytical results.) Figure 3(a) plots the zero-temperature spontaneous magnetization as a function of J_0 obtained upon slow cooling. This magnetization appears to rise abruptly from the finite-size-limited "zero" value to a nonzero value for $J_0 \sim 0.5$. This can be seen more directly in the phase diagram of Fig. 3(b), which indicates the transition from spin glass to a coexistent ferromagnetic-spin glass (or Gabay Toulouse)^{27,28} phase. Note that this critical value of J_0 closely coincides with the multicritical point. We find that the slope of the transition line from spin glass to the Gabay Toulouse state is approximately vertical at low T so that there does not appear to be any re-entrant behavior for the isotropic case. This result is consistent with recent analytical calculations²⁸ based on the infinite-range isotropic Heisenberg Hamiltonian.

B. Uniaxial anisotropy

The introduction of anisotropy produces several interesting effects.^{13,29} Not surprisingly, once anisotropy is introduced, we find that the temperature dependences of M^{fc} and M^{zfc} are qualitatively similar to those found for the Ising model (Fig. 1).

In Fig. 4(a) are plotted $Q_{||}$ and Q_{\perp} as functions of temperature for various uniaxial anisotropy constants $D > 0$.

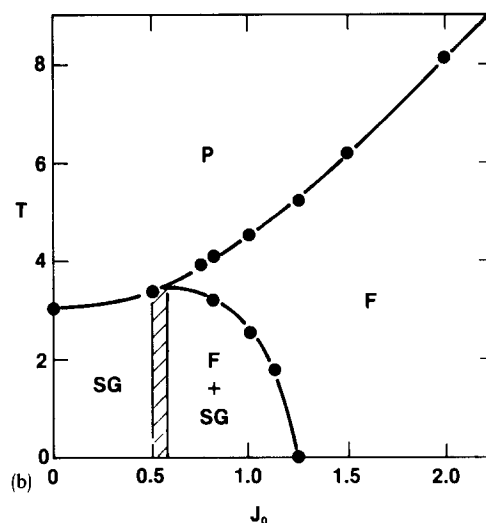
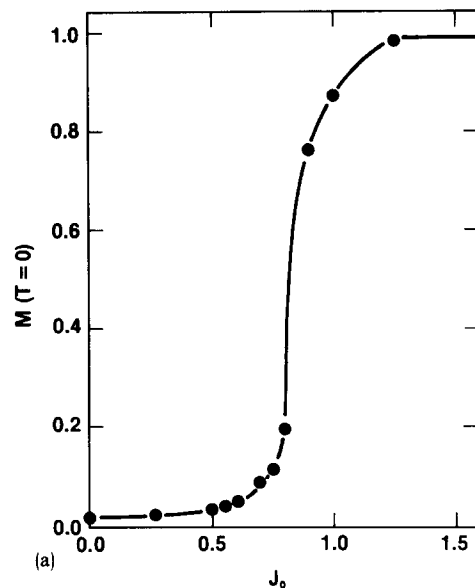


FIG. 3. (a) Zero-temperature spontaneous magnetization as a function of J_0 for the isotropic, short-range Heisenberg model for $N = 20^3$ and $S = 1$, obtained on cooling from high $T (> T_c)$ for each value of J_0 . (b) Phase diagram for this model, showing paramagnetic (P), spin glass (SG), ferromagnetic (F), and coexistence regions as a function of J_0 . The cross-hatched region indicates our uncertainty in the position of the phase boundary.

For positive D the higher transition corresponds to $Q_{||}$, whereas for $D < 0$, Q_{\perp} orders first. The phase diagram for the transverse and longitudinal spin glass transitions is shown in Fig. 4(b). Our finite-range result is qualitatively similar to that derived analytically for a classical Heisenberg spin glass with infinite-range interactions.²⁹ It should be noted that our estimates of the temperature onset for Q_{\perp} are somewhat inaccurate, due to numerical difficulties. We found no evidence for re-entrant phenomena for the uniaxial case.

C. Dzyaloshinsky moriya anisotropy

The presence of DM anisotropy,³⁰ like the uniaxial case, leads to irreversibility.¹³ The fc and zfc magnetizations are no longer macroscopically identical for nonzero values of the anisotropy constant D' .

There is a strong "competition" between DM anisotropy and a positive J_0 , or ferromagnetic bias. The former forces the spins to be at right angles, whereas the latter leads to a

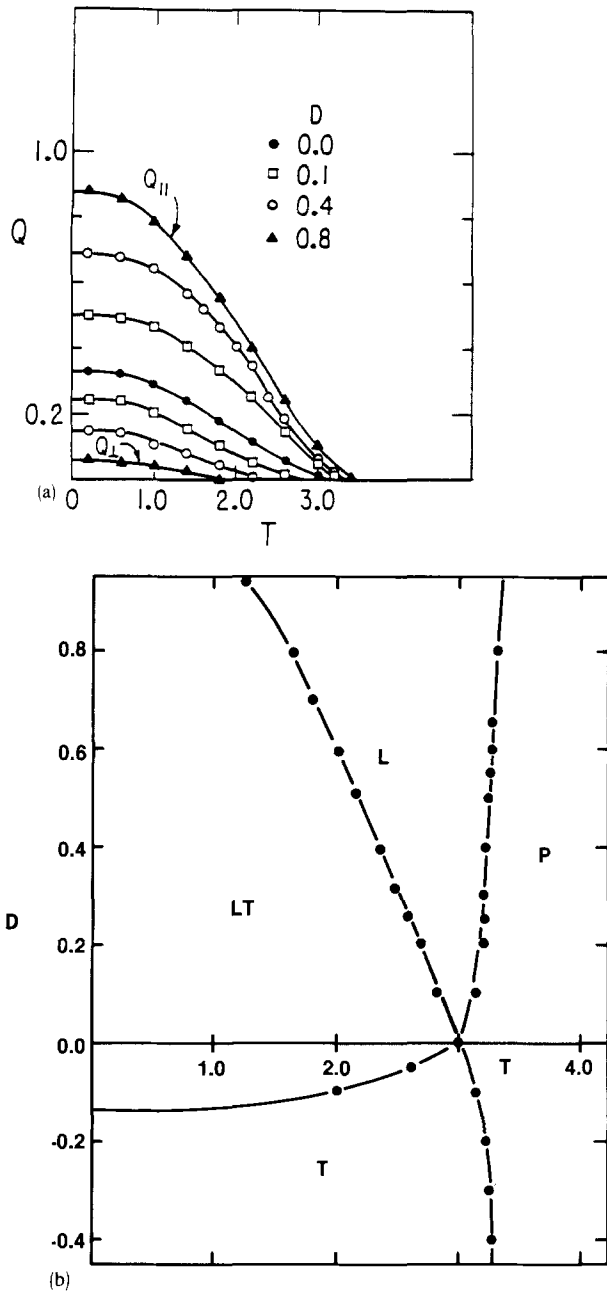


FIG. 4. (a) Temperature dependence of spin glass order parameters ($Q_{||}$ and Q_{\perp}) for different values of uniaxial anisotropy with $N = 6^3$ and $S = 1$. (b) Phase diagram for the transverse (T) and longitudinal (L) spin glass transitions in the presence of uniaxial anisotropy.

parallel alignment of the spins. Because of this competition we presumed that a Heisenberg system with DM anisotropy (and positive J_0) may exhibit re-entrant behavior. For this reason we undertook a detailed study of a spin glass system consisting of 10^3 Heisenberg spins with $D' = 0.5$.

In Fig. 5 are plotted Q_{\perp} and $Q_{||}$ as functions of T for various J_0 . Note that in contrast to the isotropic and uniaxially anisotropic cases, there is only one transition temperature for pure DM anisotropy, and $Q_{||}$ and Q_{\perp} become non-zero at the same temperature. This derives from the fact that the anisotropy contains a cross product $\mathbf{S}_i \times \mathbf{S}_j$ which ensures that Q_{\perp} is always present when $Q_{||}$ is. For this reason there can be no pure ferromagnetic phase. The presence of

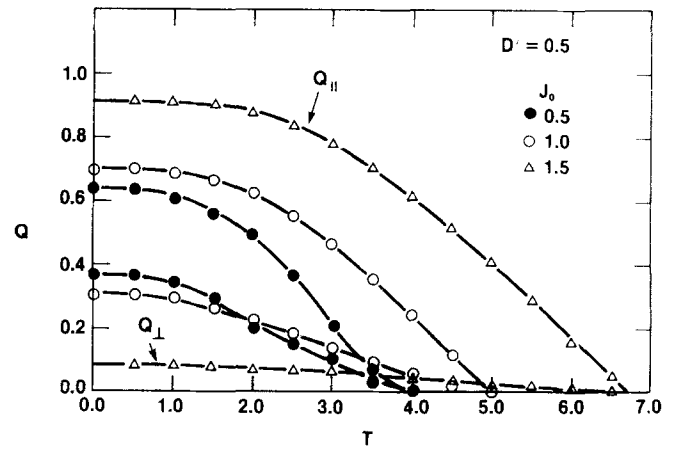


FIG. 5. Temperature dependence of the spin glass order parameters for the case of DM anisotropy for various values of J_0 for $N = 10^3$ and $S = 1$.

DM anisotropy always leads to some transverse spin glass freezing as well as longitudinal ferromagnetic order.

Figure 6 shows the phase diagram for $D' = 0.5$. For $J_0 \leq 0.6$ the low-temperature phase is a spin glass. For $J_0 \geq 0.9$ the low- T phase is a coexistent spin glass ferromagnet. For intermediate values of J_0 we were unable to find a solution to our mean field equations for a range of intermediate temperatures. Evidently the competition between J_0 and DM anisotropy is strong enough so as to totally "confuse" the system. The failure to find a converged state is associated with the "frustration" of all the spins in the system and does not reflect the behavior of a few isolated clusters. In summary, it appears to be in the most favorable region for re-entrancy that our numerical scheme has failed to follow a free energy minimum, in a slow cooling procedure. It is not obvious if this result is physical or is a consequence of our numerical scheme. It is clear that when re-entrant phenomena occur there must be a delicate balance between ferromagnetic tendencies and random exchange interactions. It is just in this region of parameter space that the solution to our equations has eluded us.

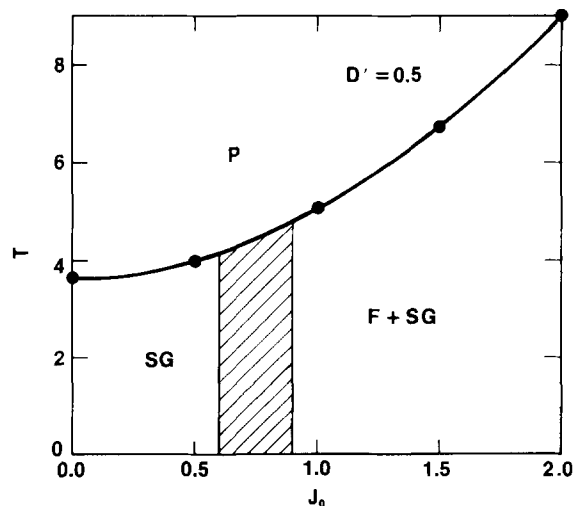


FIG. 6. Phase diagram for the short-range Heisenberg model in the presence of DM anisotropy $D' = 0.5$. In the shaded region, $0.6 < T_0 < 0.9$, we were unable to find a solution of our mean field equations. Note there is no pure ferromagnetic phase in the presence of DM anisotropy.

VI. CONCLUSIONS

The most striking conclusion of our studies is that a variety of experimental measurements can all be reasonably well reproduced by a theoretical model which focuses only on the properties of the free energy surface. Presumably, the reason for the success of this approach is that there are two important time scales in spin glass experiments: "fast times" (which may correspond to $t < 10^{-11}$ sec), during which the system makes small adjustments to find the nearest minimum of the free energy, and "long times," during which the system finds its way over energy barriers to more stable states. These latter processes appear to vary as $\ln t$, suggestive of thermal activation. It is primarily because of these long time processes that our quasistatic or quasiequilibrium viewpoint has some validity. The fast-time "dynamics" are automatically included in our calculations.

Our results for the Heisenberg model show there is no irreversibility in the isotropic, short-range Heisenberg model. This is a consequence of the ready accessibility, due to rotational symmetry, of the field-cooled state. Once microscopic anisotropy is introduced, most history-dependent properties are found to be similar to those we found for the Ising case. As reported elsewhere³¹ we have found evidence that the *infinite*-range Sherrington-Kirkpatrick model for isotropic Heisenberg spin glasses is irreversible. For this model, both the number of minima and, more importantly, the size of the barriers between them increase very rapidly with N . Thus it becomes more difficult for the system to minima hop and the system shows irreversibility.

ACKNOWLEDGMENT

This work was supported in part by the NSF under grant No. DMR 79-24007.

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